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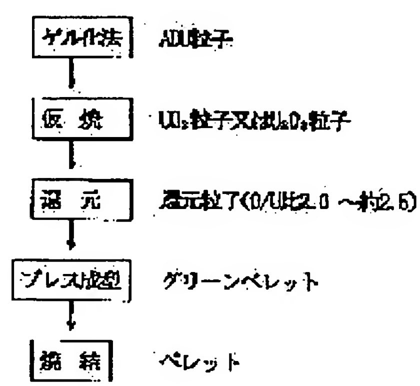
(54) PRODUCING METHOD FOR NUCLEAR FUEL PELLET USING URANIUM OXIDE PARTICLE AS RAW MATERIAL

(57)Abstract:
PROBLEM TO BE SOLVED: To prevent scattering of raw-material in producing nuclear fuel pellets, facilitate handling, improve sintering characteristics and suppress the sintering temperature low.
SOLUTION: Ammonium biuramate particles produced in an external gelation method are temporarily sintered to make UO_3 or U_3O_8 particles with the diameter of 0.1 to 1.5mm. Then, UO_2 reduced particles obtained from this are press- molded to form green pellets. By sintering the green pellets at ca. 1400° C or higher, UO_2 pellets are obtained.



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CLAIMS

[Claim(s)]
[Claim 1] Temporary quenching of the heavy uranic acid ammonium particle generated by the external gelling method is carried out at about 350-700 degrees C in air. UO3 Or U3 O8 It makes with a particle and, subsequently they are these [UO3]. Or U3 O8 A particle is returned at about 300-900 degrees C, and particle size is about 1 / 10 - 1/100 of a molding metal mold bore. UO2 It considers as a reduction particle. Furthermore, it is this UO2. It is UO2 by carrying out press molding of the reduction particle, forming the Green pellet, and sintering this Green pellet above about 1400 degrees C. The manufacture approach of the nuclear fuel pellet which uses as a raw material the uranic acid ghost particle characterized by making with a pellet.
[Claim 2] It is the above-mentioned heavy uranic acid ammonium particle, using temperature at the time of the above-mentioned temporary quenching as about 350-590 degrees C UO3 The manufacture approach of the nuclear fuel pellet which uses a particle and the uranic acid ghost particle according to claim 1 to make as a raw material.
[Claim 3] It is the above-mentioned heavy uranic acid ammonium particle, using temperature at the time of the above-mentioned temporary quenching as about 590-700 degrees C U3 O8 The manufacture approach of the nuclear fuel pellet which uses a particle and the uranic acid ghost particle according to claim 1 to make as a raw material.
[Claim 4] The gas which mixed hydrogen for the ambient atmosphere at the time of the above-mentioned reduction at inert gas, such as nitrogen, or the manufacture approach of the nuclear fuel pellet which uses as a raw material the uranic acid ghost particle according to claim 1, 2, or 3 which carried out to hydrogen gas itself.
[Claim 5] The manufacture approach of claims 1, 2, and 3 which made temperature at the time of the above-mentioned reduction 350 degrees C or more, or the nuclear fuel pellet which uses an uranic acid ghost particle given in four as a raw material.
[Claim 6] The above-mentioned heavy uranic acid ammonium particle is made to contain rare earth elements, such as Pu, TRU nuclear species, and/or FP, and it is said UO2 about each [these] element. The manufacture approach of claims 1, 2, 3, and 4 mixed in a pellet, or the nuclear fuel pellet which uses an uranic acid ghost particle given in five as a raw material.

[Translation done.]

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] This invention is not uranic acid ghost powder, and relates to the manufacture approach of the nuclear fuel pellet which used the uranic acid ghost particle as the raw material.
[0002]
[Description of the Prior Art] A nuclear fuel pellet consists of a ceramic object which usually presents the shape of a short cylinder, and where it loaded with a large number in the shape of a serial and they are sealed in a fuel rod, it is loaded in a reactor.
[0003] The above-mentioned nuclear fuel pellet is UO2 conventionally. Press molding of this powder is carried out by using powder as a raw material, and they are a consistency 5.2 - 6.0 g/cm3. They are consistency about 10.4 g/cm3 by sintering the Green pellet (green compact), and nothing and this Green pellet at an about 1700-1750-degree C elevated temperature. UO2 It is manufactured as a pellet.
[0004]
[Problem(s) to be Solved by the Invention] By the way, although the powder of an uranic acid ghost is used as a raw material like the above by the manufacture approach of the above-mentioned conventional pellet Although this powder is a massive thing which is usually several 10 - 100 micrometers of numbers which the particle 1 micrometer or less combined gently Since the diameter of powder from the first is as minute as the above-mentioned 1 micrometer or less, are hard to turn the handling by the machine up, and It may disperse in a manufacture process, and the problem of the ingredient loss and the exposure by scattering of this powder becomes serious especially, when rare earth elements, such as Pu, and TRU nuclear species or FP, are mixed in the above-mentioned raw material.
[0005] Moreover, by the above-mentioned conventional manufacture approach, in order to maintain a final consistency highly like the above-mentioned from planning oxidation stability of the powder in the middle of manufacture, sintering temperature must be made high said 1750-degree-C order.
[0006] It is made that the actual condition like the above statement should be coped with, and is not powder as an uranic acid ghost of a raw material, but this invention is UO2 of predetermined particle size. It aims at aiming at scattering prevention of the above-mentioned raw material and an improvement of a sintering property by using a particle.
[0007]
[Means for Solving the Problem] Namely, the manufacture approach of the nuclear fuel pellet of this invention which suits the above-mentioned purpose Temporary quenching of the heavy uranic acid ammonium particle generated by the external gelling method is carried out at about 350-700 degrees C in air. UO3 Or U3 O8 It makes with a particle and, subsequently they are these [UO3]. Or U3 O8 A particle is returned at about 300-900 degrees C, and particle size is about 1 / 10 - 1/100 of a molding metal mold bore. UO2 It considers as a reduction particle. Furthermore, it is this UO2. It is UO2 by carrying out press molding of the reduction particle, forming the Green pellet, and sintering this Green pellet above about 1400 degrees C. It is characterized by making with a pellet.
[0008] Moreover, it is the above-mentioned heavy uranic acid ammonium particle, using temperature at the time of the above-mentioned temporary quenching as about 350-590 degrees C in the manufacture approach of the nuclear fuel pellet of above-mentioned this invention UO3 It is the above-mentioned heavy uranic acid ammonium particle, using temperature at the time of temporary quenching as about 590-700 degrees C a particle, nothing, or the same U3 O8 Making with a particle is also possible respectively.
[0009] Use the ambient atmosphere at the time of said reduction as the gas which mixed hydrogen in inert gas, such as nitrogen, or hydrogen gas itself still more nearly similarly, and make temperature at the time of said reduction into 350 degrees C or more further, and said heavy uranic acid ammonium particle is made to contain rare earth elements, such as Pu, TRU nuclear species, and/or FP, and it is said UO2 about each [these] element. Mixing in a pellet is also possible respectively.
[0010]
[Function] In the manufacture approach of above-mentioned this invention, since the uranic acid ghost of a raw material is a particle more than the diameter of predetermined, while the handling by the machine becomes easy and it is suitable for remote operation or automation, since the above-mentioned particle cannot disperse easily compared with the conventional powder, there is very little loss in a production process, and it is effective also in contaminated reduction.

[0011] Moreover, activity is controlled for the stability reservation on the handling of raw material powder, and, for this reason, sintering temperature serves as about 1700-1750 degrees C and an elevated temperature, but the pellet which used the conventional uranic acid ghost powder as the raw material At this invention, since there is no need, a sintering property becomes good, sintering temperature of a pellet is made by this lower than said about 1400-1600 degrees C and former, and especially stabilization is UO2 of said necessary consistency. It is possible to manufacture a pellet. In addition, it is also possible to sinter it even around 1750 degrees C as usual.
[0012]
[Example] With reference to an accompanying drawing, the example of this invention is explained further below.
[0013] The flow chart Fig. in which drawing 1 shows the manufacture approach of the nuclear fuel pellet of this invention example, and drawing 2 are UO3. A particle and U3 O8 It is the graph which shows the reduction reaction of a particle, respectively.
[0014] In this invention, a heavy uranic acid ammonium particle (henceforth an ADU particle) is first generated by the gelling method.
[0015] There are the external gelling method which drop-izes the uranyl nitrate which added the gelation assistant, and particle-izes it in aqueous ammonia, and the internal gelling method which drop-izes the uranyl-nitrate undiluted solution mixed with the hexamethylenetetramine, the urea, etc., and particle-izes it in a silicone oil 100 degrees C or more as this gelling method. However, since the internal gelling method was unsuitable for industrialization in respect of generating of the ammonia of a under [gelation by low-temperature storage or the elevated temperature of mixed liquor], removal of the oil after particle-izing, etc., it generated the ADU particle by the external gelling method also in this invention.
[0016] Next, temporary quenching of the ADU particle is carried out in air, and it is UO3. A particle or U3 O8 It considers as a particle.
[0017] Since pyrolysis removal of the organic material which temporary-quenching temperature has added in an ADU particle at less than 350 degrees C is inadequate, it is UO3 in the range of about 400-550 degrees C suitably. It considers as a particle.
[0018] On the other hand, it is UO3 when temporary-quenching temperature exceeds about 590 degrees C. A particle is U3 O8. It becomes a particle. However, above 700 degrees C, it is U3 O8. A particle becomes weak. Therefore, preferably, if temporary-quenching temperature is the range which is about 400-650 degrees C, it is suitable UO3 for reduction of degree process. A particle or U3 O8 A particle can be manufactured.
[0019] And at this invention, it is the above UO3 further. Or U3 O8 Although it returns to a particle, it is the above UO3 to drawing 2 . And U3 O8 The result to which the particle was made to return in ammonolysis gas, respectively is shown. In addition, as reducing atmosphere, what added hydrogen to each inert gas, such as nitrogen, an argon, and helium, several % to about 99%, and hydrogen gas itself can be used.
[0020] UO3 The O/U ratio of a particle is 3 and a rapid reduction reaction starts at about 350 degrees C. A reduction reaction is ended at about 450 degrees C, and the O/U ratio of this reduction particle is set to 2. On the other hand, U3 O8 The O/U ratio of a particle is 2.67 and a reduction reaction starts at about 350 degrees C. A reduction reaction is ended at about 600 degrees C, and the O/U ratio of this reduction particle is set to 2. Although an O/U ratio maintains 2 [at least 600 degrees C or more], since sintering of these reduction particles begins from about 900 degrees C, both are unsuitable to the press of degree process.
[0021] Next, although press molding of the above-mentioned reduction particle is performed, the suitable reduction particle for press molding is the O/U ratio 2-2.5 They are the range and a consistency 4 - 6 g/cm3 What was made into the range is desirable. adjusting reduced temperature, as shown in drawing 2 -- UO3 a particle -- be -- U3 O8 a particle -- be -- it is alike, respectively and the O/U ratio of a reduction particle can be adjusted to arbitration. However, since there is an increment in some O/U ratio at the time of fetch, it is necessary to decide fetch conditions suitably. The degree of sintering of the O/U ratio of a reduction particle when the one [a little] higher than 2 carries out pellet molding improves.
[0022] moreover -- although it is desirable to adjust to bimodal particle size distribution from which the diameter of a reduction particle is 1/10 or less [of a dice bore], and the path of the 1st peak moreover becomes 1/10 of the paths of the 2nd peak in particle size distribution generally in order to be densely filled up with a reduction particle in a dice at the time of press molding -- a single -- even when it is modal, it does not interfere.
[0023] For example, it is filled up with a reduction particle in a dice, and is 5000kg/cm2. If pressed by the above molding pressure force, it will be consistency about 5.5 g/cm3. The Green pellet can be cast. in addition -- usually -- the inside diameter of the molding metal mold of the pellet for nuclear fuel -- about 10mm -- it is -- 1 / 10 - 1/100 of this bore it is -- the particle size of a reduction particle serves as the range of about one to 0.1 mm. Moreover, UO3 U3 O8 In order that a particle may cause about 30% of contraction at the time of reduction, the particle diameter of the particle before these reduction serves as about 1.5 - 0.1 mm.
[0024] And this Green pellet is made to sinter at about 1500 degrees C suitably in about 1400-1750 degrees C among reducing atmosphere in the last sintering process, and it is consistency 10.45 g/cm3. Considering as the above pellet is ******. The O/U ratio of a reduction particle is 2-2.5. Even if it is the range, finally after the completion of sintering, the O/U ratio of a pellet is set to 2.
[0025] (Example 1) The ADU particle was manufactured by the external gelling method. That is, PVA (polyvinyl alcohol) and 4HF (tetrahydrofurfuryl alcohol) were mixed as an additive to the uranyl nitrate, and the dropping undiluted solution was prepared. The presentation of a dropping undiluted solution was made into uranium concentration 1 mol/l, PVA30 g/l, and 4HF30vol%. The oscillating nozzle generated the drop of a dropping undiluted

solution, this was made to gel in 20wt(s)% aqueous ammonia, and it considered as the ADU particle. It controlled in order to consider as an ADU particle with a diameter of about 1mm by the oscillating nozzle.
[0026] As a temporary-quenching process, temporary quenching of the ADU particle is carried out at 550 degrees C among air, and it is UO3. It considered as the particle. Temporary quenching is carried out in an about 590-650-degree C temperature requirement, and it is U3 O8. There was especially no problem also as a particle. UO3 which carried out temporary quenching The diameter of a particle was about 0.9 mm.
[0027] As a reduction process, reduction is concentration 10wt%H2 / N2. It carries out at 440 degrees C among gas for 2 hours, and is N2. After cooling in gas, it took out at the room temperature. the O/U ratio of this reduction particle -- 2.1 it is -- particle diameter was about 0.7 mm.
[0028] The bore phi10mm dice was filled up with the above-mentioned reduction particle of about 0.7 diameter mm as a press molding process. The particle filling factor at this time was about 61 vol(s)%. It is this 6000kg/cm2 Press molding is carried out by the molding pressure force, and they are the outer diameter of 10mm, height of 10mm, and consistency about 5.7 g/cm3. It considered as the cylinder-like Green pellet. It is the above-mentioned Green pellet as a sintering process 7.5wt %H2 / N2 It sinters at 1500 degrees C among reducing atmosphere for 3 hours, and is consistency 10.45 g/cm3. The pellet was obtained.
[0029] (Example 2) Temporary quenching of the same ADU particle as said example 1 is carried out in 600-degree C air, and it is U3 O8. The particle was obtained. This U3 O8 The particle was maintaining the ball type. Furthermore, it is U3 O8. It is a particle 10wt%H2 / N2 N2 after returning to 580-degree-C ** among gas for 2 hours When it cooled in gas and having been taken out at the room temperature, the O/U ratio of this reduction particle was 2.05, and particle diameter was about 0.7 mm. Press molding was carried out by the same approach as an example 1 using this reduction particle, and the Green pellet of consistency 5.5 g/cm3 was obtained. And this Green pellet is too sintered on the same conditions as an example 1, and it is consistency 10.40 g/cm3. The pellet was obtained.
[0030]
[Effect of the Invention] As explained above, the manufacture approach of the nuclear fuel pellet of this invention Temporary quenching of the ADU particle generated by the external gelling method is carried out in air, and it is UO3 of particle size 0.1 [about] - 1.5 mm. Or U3 O8 A particle and nothing. Subsequently, these [UO3] Or U3 O8 A particle is returned and it is UO2. It considers as a reduction particle. Furthermore, it is this UO2. It is UO2 by carrying out press molding of the reduction particle, forming the Green pellet, and sintering this Green pellet. Since it makes with a pellet and the uranic acid ghost of a raw material is a particle more than the diameter of predetermined, While the handling by the machine becomes easy and it is suitable for remote operation or automation Since the above-mentioned particle cannot disperse easily compared with the conventional powder, there is very little loss in a production process. It is effective also in contaminated reduction, therefore when addition content of the rare earth elements, such as Pu, TRU nuclear species, or FP, is carried out, are advantageous. Furthermore, it receives that activity was controlled by the conventional manufacture approach for stability reservation of raw material uranium powder, and sintering temperature had become about 1700-1750 degrees C and an elevated temperature. At this invention, since it is unnecessary, a sintering property becomes good, about 200 degrees C or more of sintering temperature of a pellet are made lower than before by this, and especially stabilization is UO2 of a necessary consistency. Remarkable effectiveness that a pellet can be manufactured in energy saving is done so.

[Translation done.]

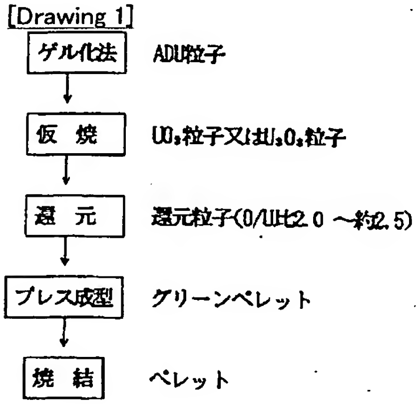
DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]
[Drawing 1] It is the flow chart Fig. showing the manufacture approach of the nuclear fuel pellet of this invention example.
[Drawing 2] UO3 A particle and U3 O8 It is the graph which shows the reduction reaction of a particle, respectively.

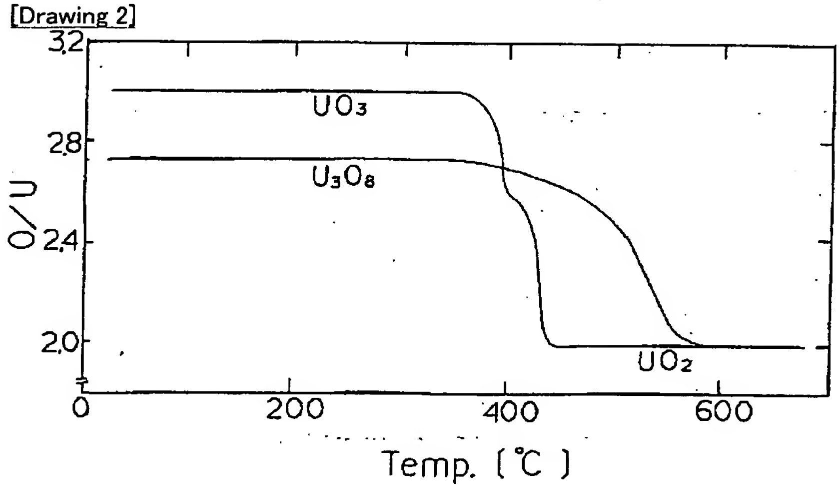
[Translation done.]

DRAWINGS

[Drawing 1]



[Drawing 2]



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